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# DETERMINATION OF RADIONUCLIDE CONCENTRATION IN MILK SAMPLES CONSUMED IN REPUBLIC OF NORTH MACEDONIA AND POPULATION DOSE RATE ESTIMATES

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**Abstract**

Milk is one of the most important food products in the human diet and contains all the macronutrients, that are, proteins, carbohydrates, fat, vitamins (A, D and B groups) and trace elements, especially calcium, phosphorus, magnesium, zinc and selenium. Milk contamination is largely due to the grazing of animals on contaminated grass and drinking water. Grass is a direct source or route of radionuclides to animals and humans through the consumption of meat and milk. One of the important tasks of the veterinary activity is veterinary-sanitary supervision of the production and sale of milk and dairy products, whose main goal is the provision of biologically good milk and dairy products from healthy animals. The purpose of this study was to determine the activity concentrations of $^{226}$Ra, $^{232}$Th $^{40}$K and $^{137}$Cs in milk samples most commonly used in daily consumption in the Republic of North Macedonia and based on the results, the risk of radiation to the population can be estimated. An instrument - gamma spectrometer (Canberra Packard) with a high purity germanium detector and GENIE 2000 programme was used for measurement of the samples. On the basis of the performed tests, the main activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K were 1.76 ± 0.23; 1.05 ± 1.00; 31.9 ± 5.07 (Bq·kg$^{-1}$), respectively. $^{40}$K has the highest value compared to other radionuclides due to the process of transfer from soil to grass and from grass and water to milk. The activity of $^{137}$Cs is below the detection limit for all tested milk types. This shows that there is no risk of radiation to the population, i.e., the safety limits are not exceeded, which points out the insignificant threat of radiation arising from radionuclides that are naturally or artificially present in the tested milk, and that reach humans through the food chain.

**Key words:** *radioactivity; milk; gamma spectrometry; radiation risk*

## INTRODUCTION

Measurements of radioactivity in the environment and food products are extremely important for controlling the levels of radiation that humans are directly or indirectly exposed to. Milk is one of the basic food products for the human diet and it contains most of the macronutrients, that is, proteins, carbohydrates, fats, vitamins (A, B and D groups) and trace elements such as calcium, phosphorus, magnesium, zinc and selenium (Kanai et al., 2013; Vreman et al.,1989). Milk contamination is largely due to the grazing of animals on contaminated grass and the drinking water they consume. Grass is a direct source or route of radionuclides to animals and humans through the consumption of meat and milk. One of the key roles of veterinary medicine is the veterinary-sanitary supervision of the production and sale of milk and dairy products, whose main goal is to provide biologically good milk and dairy products from healthy animals. If milk samples contain high levels of radioactivity when they reach the person who uses them in the daily diet, they can accumulate in certain parts of the body (radium-226 is accumulated in the lungs and kidneys, thorium-232 is accumulated in the human liver, skeleton, tissue, and lungs, while potassium-40 is usually accumulated in the muscles). Due to the presence of these radionuclides in all vital organs of the human

body, health problems may emerge that can cause various forms of diseases and weakening of the immune system and contribute to the increase in the mortality rate. The radionuclides $^{134}$Cs, $^{137}$Cs, $^{131}$I, $^{89}$Sr, $^{90}$Sr are of most interest when it comes to milk, which is a food product in the daily diet (IAEA 295, 1989) and considering the fact that they have different decay rates, the doses of these elements will be different. Therefore, monitoring the concentrations of radionuclide activity will provide important information that can contribute to knowledge about the exposure of the population and the establishment of the original baseline. This study copes with the research, i.e., determination of the activity concentrations of $^{232}$Th, $^{226}$Ra, $^{40}$K and $^{137}$Cs in milk samples that are available in our markets, i.e., in the Republic of North Macedonia. The results of the measurement of the activity concentration will be used to calculate several parameters that are important for radiological risk assessment, and the data can be used to determine the baseline for natural and artificial radioactivity in milk.

## MATERIAL AND METHODS

### Sampling

In order to collect samples of milk that is part of the dairy products consumed in the Republic of North Macedonia, a survey was conducted during 2021, which includes analysis from supermarkets, fast food restaurants and large milk distributors throughout the country. The research showed that there are about 8 leading brands of liquid milk that were consumed by a large part of the population in the Republic of North Macedonia. Two specimens were taken from each sample within the envisaged months for taking samples, in fact, different UHT milk types from a total of 8 producers were included. The samples were placed in 0.5 L Marinelli containers which were fully filled, sealed and stored in order to establish a balance between $^{226}$Ra and $^{222}$Rn before the measurements were made.

### Instrument

The research is focused on determining the level of radioactivity in raw milk by using a high-resolution HPGe detector. The gamma ray spectrometry technique was used for radioactivity determination of the tested samples. The spectrometer consisted of an HPGe detector, model 3020 (Canberra Packard, Meriden, CT, USA), with active volume of 180 cm$^2$, relative efficiency of 30 %, operating voltage 3000 V, and resolution of 2 keV at 1332.5 keV. The detector was enclosed in massive 12 cm thick lead shielding and internal lining of 2 mm high purity cooper. Data acquisition and analysis were performed with 8192 channel digital analyzer; duration of acquisition interval for each sample was 65 ks. The activity of $^{226}$Ra was determined from the gamma lines associated with low half-live time daughters of 214Bi (609.31, 1120.29, and 1794.49 keV) and $^{214}$Pb (351.93 keV). The $^{232}$Th activity was determined by 338.4, 911.2 and 969.1 keV gamma lines form 228Ac and its decay products. The gamma line at 1460.8 keV was used to determine the activity of 40K.Efficiency calibration was performed with mixed calibration standard sources MBSS2, supplied from the Czech Metrological Institute, Inspectorate for Ionizing Radiation. In order to determine the background distribution in the detector environment, empty sealed Marinelli beaker with the same geometry was measured at equal counts as the soil samples. The analysis procedure included the subtraction of the background spectrum.

### -Activity calculation

The specific activity (A) is determined according the equation

$$A = \frac{\frac{N}{t} - \frac{N_0}{t_0}}{\varepsilon \cdot \gamma \cdot m} \quad (Bq \cdot kg^{-1})$$

Where, N is clean surface of peak accumulated from a specific radionuclide in analysis of a specific sample (number of readings), $N_0$ is clean surface of peak accumulated from the spot of a specific radionuclide without an analysis of sample (number of readings), t is live time of accumulation of the sample spectrum (s), $t_0$ is live time of accumulation of the phone spectrum (s), ε is detector efficiency for a given energy (for a specific peak), γ is intensity of gamma transition in radioactive decay for a respective radionuclide (%), and m is sample mass (kg).

### -Air absorbed dose rate (D)

A direct connection between radioactivity

concentrations of natural radionuclides and their exposure is known as the absorbed dose rate in the air at 1 meter above the ground surface. The mean activity concentrations of $^{232}$Th, $^{226}$Ra and $^{40}$K(Bq·kg$^{-1}$) in the samples are used to calculate the absorbed dose rate given by the following formula. (Belivermis et al., 2010) D (nGy / h) = 0,462 $A_{Ra}$ + 0,604 $A_{Th}$ +0,0417 $A_k$ +0,030 $A_{Cs}$

**-Radium equivalent activity (Raeq)**

The model of the radium equivalent activity establishes the use of a single index to define the gamma output or compare the specific activities of materials containing $^{226}$Ra, $^{232}$Th, and $^{40}$K by a single quantity, which takes into consideration the radiation risk associated with these NORMs (Roy et al., 2000). The calculation of the radium equivalent activity (Raeq) is a quantity for comparing the specific activities of the samples with different contents of $^{226}$Ra, $^{232}$Th and $^{40}$K. The uniformity with respect to radiation exposure was defined in terms of the radium equivalent activity (Raeq) in Bq·kg$^{-1}$ in order to compare the specific activity of the materials containing different amounts of $^{226}$Ra, $^{232}$Th and $^{40}$K. It is assumed that 370 Bq·kg$^{-1}$ of $^{226}$Ra, 259 Bq·kg$^{-1}$ of $^{232}$Th and 4810 Bq·kg$^{-1}$ of $^{40}$K produce the same gamma-ray dose rate. It is calculated by using the following ratio $Ra_{eq}$ (Bq/kg) = $A_{Ra}$+1.43$A_{Th}$+0.07 $A_k$ (Beretka et al., 1985) $A_{Ra}$, $A_{Th,}$ $A_k$ – specific activities (Bq·kg$^{-1}$) of $^{226}$Ra, $^{232}$Th and $^{40}$K, respectively. The value of the radium equivalent activity of 370 Bq·kg$^{-1}$ corresponds to the maximum allowed dose for a population of 1 mSv.

**-External and internal hazard index**

In order to assess the equivalent average of the annual effective dose imposed to the residents of each area, the external hazard index for the soil samples was calculated.

$$H_{eks} = A_{Ra}/370 + A_{Th}/259 + A_k/4810 \leq 1$$

$A_{Ra}$, $A_{Th,}$ $A_{k-}$specific activites (Bq·kg$^{-1}$) , $^{226}$Ra$^{232}$ Th and $^{40}$K, respectively (Kurnaz et al., 2007).

**-Annual effective dose equivalent (AEDE)**

The annual effective dose equivalent received was computed from absorbed dose rate by applying a dose conversion factor of 0.7 Sv Gy−1 and the occupancy of 0.8 (19/24) recommended by UNSCEAR. Therefore, the annual effective dose equivalent (μS vy−1) was calculated using the formula (UNSCEAR., 2000)

AEDE(μSvy−1) = absorbed dose (nGyh−1)×8760h×0.7SvGy−1×0.8×10−3

- **Excess lifetime cancer risk**

Excess life cancer risk predicts the likelihood of developing cancer over a lifetime at a certain exposure rate. It is a value representing the number of extra cancers expected in a given number of people on exposure to a carcinogen at a given dose.

Excess lifetime cancer risk is given as (Taskin et al., 2009)

ELCR=AEDE×DL×RF

The parameters used are defined; thus, AEDE is the annual effective dose equivalent, DL is the average duration of life (estimated to be 70 years), and RF is the risk factor (S/v), i.e., fatal cancer risk per Sievert. ICRP uses a RF of 0.05 for the public for stochastic effects.

If a person consumes milk that contains elevated levels of radionuclides, this may increase the chances of cancer. If the radioactivity in milk is higher than the world average, this could be a source of radiation to the human body and some specific organs, whereby their ELCR would be higher than the world average of 0.29 mS vy-1 in such a body.

## RESULTS AND DISCUSSION

The activity concentrations of $^{232}$Th, $^{226}$Ra, $^{40}$K and $^{137}$Cs in selected brands of fresh milk are presented in Table 1.

**Table 1.** $^{232}$Th, $^{226}$Ra ,$^{40}$K and $^{137}$ Cs activity concentrations of milk in January, 2021

| | **Specific activity* (Bq·kg$^{-1}$)** | | | |
|---|---|---|---|---|
| Month | **January, 2021** | | | |
| Sample | $^{226}$Ra | $^{232}$Th | $^{40}$K | $^{137}$ Cs |
| S1 | 1.80±0.20 | 0.75±0.15 | 34.9 ± 6.20 | <MDA* |
| S2 | 1.99±0.21 | 1.18±0.15 | 32.7 ± 5.3 | <MDA |
| S3 | 1.91±0.15 | 1.26±0.20 | 31.9± 5.5 | <MDA |
| S4 | 1.65±0.16 | 0.63±0.14 | 32.0 ± 4.2 | <MDA |
| S5 | 1.22±0.18 | 1.09±0.16 | 29.1 ± 3.5 | <MDA |
| S6 | 2.52±0.15 | 1.64±0.15 | 36.2 ± 5.0 | <MDA |
| S7 | 2.44±1.00 | 1.21 ± 0.17 | 36.4±5.2 | <MDA |
| S8 | 1.11±0.50 | 0.72±0.19 | 21.0±5.5 | <MDA |
| Average | 1.83±0.31 | 1.06±0.16 | 31.0±5.05 | <MDA |

*MDA - minimum detectable activity, number of repetitions for each sample (n=3)

**Table 2.** $^{232}$Th, $^{226}$Ra ,$^{40}$K and $^{137}$Cs activity concentrations of milk in April, 2021

| | **Specific activity* (Bq·kg$^{-1}$)** | | | |
|---|---|---|---|---|
| Month | **April, 2021** | | | |
| Sample | $^{226}$Ra | $^{232}$Th | $^{40}$K | $^{137}$ Cs |
| S1 | 1.81±0.24 | 0.85±0.15 | 35.6 ± 7.80 | <MDA |
| S2 | 2.04±0.24 | 1.18±0.16 | 33.2 ± 5.21 | <MDA |
| S3 | 1.86±0.17 | 1.21±0.24 | 34.2± 5.66 | <MDA |
| S4 | 1.45±0.18 | 0.60±0.18 | 31.4 ± 4.6 | <MDA |
| S5 | 1.15±0.18 | 0.99±0.16 | 29.7 ± 3.0 | <MDA |
| S6 | 2.22±0.16 | 1.64±0.17 | 36.8 ± 4.1 | <MDA |
| S7 | 2.14±0.20 | 1.21 ± 0.17 | 37.4±6.8 | <MDA |
| S8 | 1.02±0.10 | 0.75±0.20 | 21.6±6.8 | <MDA |
| Average | 1.70±0.18 | 1.05±0.17 | 32.4±5.50 | <MDA |

*MDA - minimum detectable activity, number of repetitions for each sample (n=3)

**Table 3.** $^{232}$Th, $^{226}$Ra ,$^{40}$K and $^{137}$Cs activity concentrations of milk in August, 2021

| | **Specific activity* (Bq·kg$^{-1}$)** | | | |
|---|---|---|---|---|
| Month | **August, 2021** | | | |
| Sample | $^{226}$Ra | $^{232}$Th | $^{40}$K | $^{137}$ Cs |
| S1 | 1.82±0.21 | 0.77±0.14 | 35.0 ± 5.00 | <MDA |
| S2 | 2.24±0.24 | 1.26±0.14 | 33.4 ± 5.00 | <MDA |
| S3 | 1.97±0.15 | 1.11±0.20 | 32.1± 5.00 | <MDA |
| S4 | 1.52±0.11 | 0.68±0.11 | 31.7 ± 4.80 | <MDA |
| S5 | 1.66±0.10 | 1.29±0.17 | 29.6 ± 3.50 | <MDA |
| S6 | 2.44±0.19 | 1.52±0.15 | 36.9 ± 4.50 | <MDA |
| S7 | 2.33±1.0 | 0.99 ± 0.17 | 35.4±6.50 | <MDA |
| S8 | 1.17±0.55 | 0.75±0.18 | 20.9±6.80 | <MDA |
| Average | 1.83±0.31 | 1.04±0.15 | 31.87±4.53 | <MDA |

*MDA - minimum detectable activity, number of repetitions for each sample (n=3)

**Table 4.** $^{232}$Th, $^{226}$Ra ,$^{40}$K and $^{137}$Cs and activity concentrations of milk in October, 2021

| | **Specific activity* (Bq·kg$^{-1}$)** | | | |
|---|---|---|---|---|
| Month | **October, 2021** | | | |
| Sample | $^{226}$Ra | $^{232}$Th | $^{40}$K | $^{137}$Cs |
| S1 | 1.81±0.18 | 0.88±0.10 | 36.1 ± 6.80 | <MDA |
| S2 | 2.04±0.17 | 1.11±0.11 | 33.2 ± 5.20 | <MDA |
| S3 | 1.86±0.17 | 1.22±0.23 | 31.2± 5.30 | <MDA |
| S4 | 1.45±0.15 | 0.61±0.15 | 31.4 ± 4.50 | <MDA |
| S5 | 1.15±0.15 | 0.99±0.16 | 28.7 ± 3.00 | <MDA |
| S6 | 2.22±0.10 | 1.51±0.16 | 36.1 ± 4.00 | <MDA |
| S7 | 2.14±0.15 | 1.06 ± 0.17 | 35.9±6.50 | <MDA |
| S8 | 1.02±0.13 | 0.66±0.19 | 21.6±6.50 | <MDA |
| Average | 1.71±0.15 | 1.05±0.15 | 31.77±5.22 | <MDA |

*MDA - minimum detectable activity, number of repetitions for each sample (n=3)

**Table 5.** $^{232}$Th, $^{226}$Ra ,$^{40}$K and $^{137}$Cs activity concentrations of milk samples, mean values from the four seasons

| **Radionuclides** | **Bq·kg$^{-1}$** |
|---|---|
| 226Ra | 1.76±0.23 |
| 232Th | 1.05±1.00 |
| 40K | 31.9±5.07 |
| 137Cs | <MDA Bq·kg$^{-1}$ |

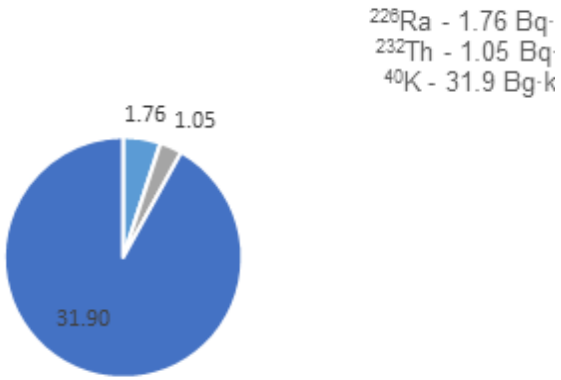


**Figure1.** Graphical representation of mean values of the three natural radionuclides measured in milk

$^{40}$K is the dominant radionuclide in the tested samples. More specifically, all samples, in all seasons, have the highest concentration of activity of $^{40}$K with an average value of 31.9 ± 5.07 Bq·kg$^{-1}$. The higher concentration of activity of $^{40}$K in milk samples can be justified since potassium is very mobile in the environment, it is one of the major radionuclides in the soil and naturally forms part of potassium, which is a major nutrient for plants, animals and humans (Hafsi et al., 2014; Bilgici Cengiz et al., 2019). These reasons lead to a high concentration of activity of $^{40}$K compared to other radionuclides due to the process of transfer from soil to grass and from grass and water to milk. The average concentration of radioactivity of $^{226}$Ra in liquid milk is 1.76 ± 0.23 Bq·kg$^{-1}$, while the average concentration of activity of $^{232}$Th is 1.05 ± 1.0 Bq·kg$^{-1}$. Actually, the value of cesium 137 is below the detection limit. The values of the analysed samples are below the permitted limits UNSEAR 2000. If the results for all radionuclides are summarized, it can be seen that there are no statistically significant differences between the radioactivity concentrations between the samples. Furthermore, the activity of the concentrations of $^{232}$Th, $^{226}$Ra, $^{137}$Cs and $^{40}$K in milk that was found in this study is compared

with reports from other authors in different countries where it can be seen that there are variations of the concentrations and the values in our study are below the obtained values for milk consumed in some countries such as Iran/France (Hosseni et al., 2006), Jordan (Zaid et al., 2010), Egypt (Harb et al., 2010), Turkey (Cengiz., 2020), Brazil (Melquiades et al., 2002) and other countries. Although the differences in the levels of radioactivity in different brands of milk are small, it is considered that the source of raw materials used for milk production is the contributing factor. Previous researches shown that the average annual value of the concentration of 137Cs activity in milk samples in Serbia in 1985 was 0.11 Bq / l. After the Chernobyl accident, its activity concentration values reached 72.6 Bq / l for cesium. Since 1987, this value has been declining exponentially, and in 2013, minimum values of 0.038 Bq / l for 137Cs were measured. (Bogojevic et al., 2016). From the obtained results shown in Tables 1,2,3 and 4, we can conclude that there is a non-significant difference in terms of specific activity for all examined radionuclides in different months of the year.

Based on the mean values of the specific activities for the radionuclides $^{232}$Th, $^{226}$Ra, $^{40}$K and $^{137}$Cs, the following values were calculated: the radium equivalent activity (Raeq), the absorbed dose rate D (nGy/h), the internal hazard indices (Hin), the received annual effective dose equivalent (AEDE) and the excess lifetime cancer risk (ELCR) for the different samples of dairy products, which are presented in Table 6.

**Table 6.** D, Raeq, Hex, AEDE, ELCR Risk of lifetime cancer values of dairy products

| Sample (Month) | D (nGy/h) | Raeq (Bq·kg$^{-1}$) | Hex | AEDE(μSv/year) | ELCR (μSv/year) |
|---|---|---|---|---|---|
| January | 2.80 | 5.51 | 0.01 | 13.73 | 48 |
| April | 2.79 | 5.46 | 0.01 | 13.68 | 47.88 |
| August | 2.83 | 5.54 | 0.01 | 13.88 | 48.58 |
| October | 2.77 | 5.43 | 0.01 | 13.58 | 47.53 |
| Average | 2.80 | 5.48 | 0.01 | 13.71 | 47.99 |

The mean value of the radiation risk index Heks is 0.01 whose value is less than the maximum allowed value which for Heks is <1. The value of the radium equivalent activity Raeq is below the maximum recommended limit, i.e., 370 Bq kg$^{-1}$ which is 5.48 Bq kg$^{-1}$. The values of the specific activity and the calculated Radiation Risk Index (Heks) and Radiation Equivalent (Raeq) obtained in this study also did not exceed the safety limits, emphasizing the insignificant radiation hazard which arises from naturally occurring terrestrial radionuclides. The values of the absorbed dose rate D (nGy/h) and the annual effective dose equivalent (AEDE) obtained in this study, also did not exceed safety limits, emphasizing the negligible radiation hazard arising from naturally present terrestrial radionuclides. The estimated ELCR obtained in all measured samples is lower than the international standard limit.

## CONCLUDING REMARKS

The gamma-spectrometry assessment of natural radioactivity in milk consumed in the Republic of North Macedonia is presented in this study. It was found that the concentration of radionuclides in milk samples is determined by the source from which the milk was obtained. The mean values of the concentration of specific activity in the milk samples were lower than the permitted public dose limit worldwide. All calculated radiological risk parameters show that none of the milk samples exceeded the recommended permitted level. From the research it can be concluded that the milk consumed in the Republic of North Macedonia is radiologically safe and cannot cause immediate or significant threat to the health of the consumers of the examined milk brands. However, continuous control of the radiological safety of milk and all dairy products consumed by the population in the Republic of North Macedonia is recommended.

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# ОПРЕДЕЛУВАЊЕ НА КОНЦЕНТРАЦИЈАТА НА РАДИОНУКЛИДИ ВО ПРИМЕРОЦИ НА МЛЕКО КОРИСТЕНИ ВО РЕПУБЛИКА СЕВЕРНА МАКЕДОНИЈА И ПРОЦЕНКИ НА РИЗИК ОД РАДИЈАЦИЈА НА НАСЕЛЕНИЕТО

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**Резиме**

Млекото е еден од најважните прехранбени производи во човековата исхрана и ги содржи сите макронутриенти, односно протеини, јаглени хидрати, масти, витамини (А, Д и Б групи) и елементи во траги, особено калциум, фосфор, магнезиум, цинк и селен. Контаминацијата на млекото во голема мера се должи на пасењето на животните на контаминирана трева и вода за пиење. Тревата е директен извор или пат на радионуклиди до животните и луѓето преку потрошувачката на месо и млеко. Една од важните задачи на ветеринарната дејност е ветеринарно-санитарниот надзор на производството и продажбата на млеко и млечни производи, чија основна цел е обезбедување на биолошки добро млеко и млечни производи од здрави животни. Целта на оваа студија беше да се утврдат концентрациите на активност на 226Ra, 232Th 40K и 137Cs во примероците на млеко кои најчесто се користат во секојдневната потрошувачка во Република Северна Македонија и врз основа на резултатите може да се процени ризикот од зрачење на населението. За мерење на примероците е користен инструмент - гама спектрометар (Canberra Packard) со детектор на германиум со висока чистота и програма GENIE 2000. Врз основа на извршените тестови, средните концентрации на активност на 226Ra, 232Th и 40K беа 1,723 ± 0; 1,05 ± 1,00; 31,9 ± 5,07 (Bq·kg-1), соодветно. 40K има најголема вредност во споредба со другите радионуклиди поради процесот на пренос од земја на трева и од трева и вода во млеко. Активноста на 137Cs е под границата за детекција за сите тестирани типови млеко. Ова покажува дека не постои ризик од зрачење за населението, односно не се надминуваат безбедносните граници, што укажува на незначителна закана од зрачење што произлегува од радионуклиди кои природно или вештачки се присутни во тестираното млеко, а кои стигнуваат до луѓето преку синџир на исхрана.

**Клучни зборови:** *радиоактивност, млеко, гама спектрометрија, ризик од зрачење*